



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re Application of
Tadashi Ishida, et al.
Application No. 10/522,416
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For: INK JET RECORDING MEDIUM

Group Art Unit: 1794
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Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

DECLARATION UNDER 37 CFR 1.132

I, Masaya KUSUMOTO, declare and state that:

1. In March, 1990, I was graduated from Hokkaido Asahikawa Technical High-School.

Since April, 1990, I have been an employee of MITSUI Chemicals, INC., and till the present time, I have been engaged in development work in emulsion for thermosensitive recording material and for ink jet recording material.

2. I am a co-inventor of the invention described in the specification of the above-identified application.

3. I calculated SP (solubility parameter) values, d , of polymers prepared in Experiments I, II and III in the declaration of December 3, 2007, by Fedors method as a guide of water solubility.

(1) The procedure for calculating SP value according to Fedors method is as follows.

<Calculation of SP value of homopolymer>

$$d = (\Delta E/V)^{1/2} = (S\Delta e_i/S\Delta v_i)^{1/2}$$

ΔE : Cohesive energy density

V : Molar volume

Δe_i : Evaporation energy of atomic group

Δv_i : Molar volume of atomic group

When T_g of polymer is not less than 25°C and n is less than 3, the following constant (i) is added to Δv_i .

When T_g of polymer is not less than 25°C and n is not less than 3, the following constant (ii) is added to Δv_i .

(i) $4n$

(ii) $2n$

n : Number of main-chain skeletal atom in a minimum repeating unit of polymer

<Calculation of SP value of copolymer>

$$d = (d_a*d_a + d_b*d_b + d_c*d_c \dots)/100$$

$d_a, d_b, d_c \dots$: Volume % of monomer

$d_a, d_b, d_c \dots$: SP value of homopolymer

(2) According to the above procedure, SP values of homopolymers formed from monomers used in Experiments I, II and III were calculated. The results are shown in Table 1.

Table 1

Monomer	Atomic group		Δe_i		Δv_i		Homopolymer	
	Type	Number (a)	(b)	(a)*(b)	(c)	(a)*(c)	Tg [°C]	SP [cal/cm ³]
Methacrylic acid CH ₂ CCH ₃ COOH (density: 1.055)	CH ₂	1	1180	1180	16.1	16.1	185	11.76
	CH ₃	1	1125	1125	33.5	33.5		
	C	1	350	350	-19.2	-19.2		
	COOH	1	6600	6600	28.5	28.5		
	4n (n=2)					8		
	total			9255		66.9		
Dimethylaminopropyl methacrylamide CH ₂ CCH ₃ CONH CH ₂ CH ₂ CH ₂ NCH ₃ CH ₃ (density: 0.9419)	CH ₂	4	1180	4720	16.1	64.4	96	10.64
	CH ₃	3	1125	3375	33.5	100.5		
	C	1	350	350	-19.2	-19.2		
	CONH	1	8000	8000	9.5	9.5		
	N	1	1000	1000	-9	-9		
	4n (n=2)					8		
2-Hydroxyethyl methacrylate CH ₂ CCH ₃ COO CH ₂ CH ₂ OH (density: 1.072)	CH ₂	3	1180	3540	16.1	48.3	80	11.96
	CH ₃	1	1125	1125	33.5	33.5		
	C	1	350	350	-19.2	-19.2		
	COO	1	4300	4300	18	18		
	OH	1	5220	5220	13	13		
	4n (n=2)					8		
Methacrylamide CH ₂ CCH ₃ CONH ₂ (density: 1.11)	CH ₂	1	1180	1180	16.1	16.1	256	15.05
	CH ₃	1	1125	1125	33.5	33.5		
	C	1	350	350	-19.2	-19.2		
	CONH ₂	1	10000	10000	17.5	17.5		
	4n (n=2)					8		
	total			12655		55.9		
Styrene CH ₂ CHC ₆ H ₅ (density: 0.9048)	CH ₂	1	1180	1180	16.1	16.1	104	10.09
	CH	1	820	820	-1	-1		
	C ₆ H ₅	1	7630	7630	71.4	71.4		
	4n (n=2)					8		
	total			9630		94.5		
t-Butylmethacrylate CH ₂ CCH ₃ COO CCH ₃ CH ₃ CH ₃ (density: 0.874)	CH ₂	1	1180	1180	16.1	16.1	107	8.81
	CH ₃	4	1125	4500	33.5	134		
	C	2	350	700	-19.2	-38.4		
	COO	1	4300	4300	18	18		
	4n (n=2)					8		
	total			10680		137.7		

(3) According to the above procedure, SP values of copolymers prepared in Experiments I, II and III were calculated.

[Experiment I]

Volume % of monomers used in Experiment I is shown in Table 2.

Table 2

Reagent Name	% mol	% wt	% vol
Methacrylic Acid	9.662	6.24	6.04
Dimethylaminopropyl Methacrylamide	30.00	35.16	38.13
2-Hydroxyethyl Methacrylate	60.00	58.60	55.83
Trimethylolpropane Trimethacrylate	0.300		
Polymerization initiator	0.038		
Total (Polymer)	100	100	100

Trimethylolpropane trimethacrylate is eliminated from calculation because of low amount.

From Tables 1 and 2, SP value, d, of the copolymer prepared in Experiment I is calculated as follows.

$$d = \{ (6.04 \times 11.76) + (38.13 \times 10.64) + (55.83 \times 11.96) \} / 100$$

$$= 11.44 \text{ [cal/cm}^3\text{]}$$

[Experiment II]

Volume % of monomers used in Experiment II is shown in Table 3.

Table 3

Reagent Name	% mol	% wt	% vol
Methacrylamide	74.362	64.04	60.49
Dimethylaminopropyl Methacrylamide	20.00	31.61	35.19
Methacrylic Acid	5.00	4.35	4.32
Methylene Bisacrylamide	0.60		
Polymerization initiator	0.038		
Total (Polymer)	100	100	100

Methylene bisacrylamide is eliminated from calculation because of low amount.

From Tables 1 and 3, SP value, d, of copolymer prepared in Experiment II is calculated as follows.

$$d = \{ (60.49 \times 15.05) + (35.19 \times 10.64) + (4.32 \times 11.76) \} / 100$$

$$= 13.36 \text{ [cal/cm}^3\text{]}$$

[Experiment III]

Volume % of monomers used in Experiment III is shown in Table 4.

Table 4

Reagent Name	% mol	% wt	% vol
Styrene	45.35	39.40	39.68
t-Butyl Methacrylate	38.33	45.45	47.38
2-Hydroxyethyl Methacrylate	9.31	10.10	8.58
Methacrylic acid	7.01	5.05	4.36
Polymerization initiator		0.30	
Total Solids	100	100	100

From Tables 1 and 4, SP value, d, of copolymer prepared in Experiment III is calculated as follows.

$$d = \{ (39.68 \times 10.09) + (47.38 \times 8.81) + (8.58 \times 11.96) \\ + (5.05 \times 11.76) \} / 100$$

$$= 9.80 \text{ [cal/cm}^3\text{]}$$

4. From the above results of SP values calculated by Fedors method, SP value of the copolymer of the present invention is lower than that of copolymers of US6,361,768. I think that the difference of water solubility between the copolymer of the present invention and the copolymer of US6,361,768 is attributed to SP value.

In addition, copolymers of US6,361,768 comprise a non-ionic hydrophilic monomer such as 2-hydroxyethyl methacrylate and methacrylamide at a high concentration (35-95 mole percent). On the other hand, the copolymer of the present invention comprises a non-ionic hydrophilic monomer at a low concentration (9.31 mole % in Experiment III). I think that the difference of SP value between the copolymer of the present invention and the copolymer of US6,361,768 is attributed to the content of a non-ionic hydrophilic monomer.

Therefore, I believe that the hydrophilic ampholytic polymers of US6,361,768 can not be dispersed in water in the state of particle because a non-ionic hydrophilic monomer is at a high concentration.

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Masaya Kusumoto
Masaya KUSUMOTO

Date: September 5, 2008